

1 Unconventional Density Waves in Organic Conductors and in Superconductors

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Unconventional density waves (UDW) are one of the ground states in metallic crystalline solids and have been speculated already in 1968. However, more focused studies on UDW started only recently, perhaps after the identification of the low temperature phase in α -(BEDT-TTF)₂KHg(SCN)₄ as unconventional charge density wave (UCDW) in 2002. More recently, the metallic phase of Bechgaard salts (TMTSF)₂X with X=PF₆ and ReO₄ under both pressure and magnetic field appears to be unconventional spin density wave (USDW). The pseudogap regime of high T_c superconductors LSCO, YBCO, Bi2212 and the one in CeCoIn₅ belong to d-wave density waves (d-DW).

In these identifications, the angular dependent magnetoresistance and the giant Nernst effect have played the crucial role. These are the simplest manifestations of the Landau quantization of quasiparticle energy in UDW in the presence of magnetic field (the Nersesyan effect). Also we speculate that UDW will be most likely found in α -(BEDT-TTF)₂I₃, α -(BEDT-TTF)₂I₂Br, κ -(BEDT-TTF)₂Cu(NCS)₂, κ -(BEDT-TTF)₂Cu(CN)₂Br, λ -(BEDT)₂GaCl₄ and in many other organic compounds.

1.1 Introduction

Until very recently, the electronic ground state in crystalline solids were considered to belong to one of four canonical ground states in quasi-one-dimensional systems: s-wave superconductors, p-wave superconductors, (conventional) charge density waves and spin density waves [1–4]. Indeed many condensates discovered since 1972 have appeared to be accommodated comfortably in this scheme. For example the two CDW in NbSe₃ and SDW in (TMTSF)₂PF₆ are well known examples [3, 4].

In all these systems, the elementary excitations are of Fermi liquid nature à la Landau [5–7] but have the energy gap Δ and the quasiparticle

(condensate) density decreases exponentially like $\exp(-\Delta/T)$ for $T \leq T_c/2$, where T_c is the transition temperature. Therefore only a small portion of the quasiparticles will be left below $T_c/2$.

The thermodynamics of these systems are practically the same as those in s-wave superconductors as described by the theory of Bardeen, Cooper and Schrieffer (the BCS theory [8]).

However, since the discovery of heavy fermion superconductors CeCu_2Si_2 [9], organic superconductors in Bechgaard salts $(\text{TMTSF})_2\text{PF}_6$ [10], high T_c cuprate superconductors [11], Sr_2RuO_4 [12] etc., this simple scheme has to be necessarily modified. First of all, most of these superconductors are unconventional and nodal [13–17]. Parallel to this development, there is a surge of studies on CDW whose quasiparticle energy gap $\Delta(\mathbf{k})$ has line or point nodes at the Fermi surface [18–21]. We may characterize this as the paradigm shift from one dimensional to higher dimensional physics [22]. However, such paradigm shift been already anticipated. The quasiparticle excitations are of Fermi liquid type and not of Tomonaga-Luttinger type [4, 23, 24]. Note that we are not interested in the high temperature behaviour $T > 200$ K of Bechgaard salts currently discussed in literature [25]. In higher dimensions, the imperfect nesting is inevitable. Further in a magnetic field, the quasiparticle spectrum is quantized á la Landau in both SDW [26] and CDW [27]. This gives rise to field induced spin density wave (FISDW) with integral quantum Hall effect [4, 28, 29].

Unconventional density waves were first speculated by Halperin and Rice [30] as a possible ground state in the excitonic insulator. Unlike conventional DW, there is no X-ray or spin signal due to the zero average of the gap over the Fermi surface ($\langle \Delta(\mathbf{k}) \rangle = 0$). Therefore UDW is often called as the state with hidden order parameter. However, certain confusion has spread around in recent literature with the use of "hidden order". Therefore it is much wiser to specify what one means. In this sense we prefer to use the notation UDW. Also unlike many other people [18, 19, 21, 31, 32], we do not consider the discretized lattice and UDW with minuscule loop current or Z_2 symmetry breaking common to the descendent of the flux phase [31, 32].

It is easy to see that such two dimensional solution is unstable in three dimensional environment. In contrast, our UDW has the $U(1)$ gauge symmetry as in conventional DW. Therefore our UDW can slide, and exhibits the nonohmic conductance [33, 34] as in conventional DW and generates the phase vortices.

As an other difference compared to conventional DW, the nodal excitations persist down to $T = 0$ K, giving rise to an electronic specific heat $\sim T^2$. Indeed, the thermodynamics of UDW is identical to the one in d-wave superconductors [20, 35]. The transition from the normal metallic state to UDW is metal-to-metal. As an example, the T^2 dependence of the electric resistance in the normal state typical to Fermi liquids changes to T linear

dependence for $T \leq T_c/2$, which is to be contrasted to the exponentially activated behaviour in the conventional counterpart.

In light of the discussion above, many of the so-called non-Fermi liquids can be UDW, and in fact Fermi liquids as defined by Landau, as we shall see. The change in the exponents distinguishing between various phases stems from the change in the excitation spectrum, and not from the different nature of the elementary excitations. We have shown already that the pseudogap phases in both high T_c cuprates like LSCO, YBCO and Bi2212 [36, 37] and CeCoIn₅ [38, 39] are d-wave density waves based on the giant Nernst effect and the angle dependent magnetoresistance observed in these systems [40–44]. Through the angle dependent photoemission spectra, the $d_{x^2-y^2}$ symmetry of the energy gap in the pseudogap region of high T_c cuprates has been established [45]. Also the presence of Fermi arc around the (π, π) direction [46] is consistent with d-DW [47]. Furthermore, the mysterious relation $\Delta(0) \simeq 2.14T^*$ found in the pseudogap region of LSCO, YBCO and Bi2212 [48–51] can readily be interpreted in terms of UDW. Here $\Delta(0)$ is the maximal energy gap determined by STM and T^* is the pseudogap crossover temperature, which may be identified as T_c in d-DW. The 2.14 gap maximum-transition temperature ratio is the weak-coupling theory value for d-DW and for d-wave superconductors [20, 35].

1.2 Mean-field theory

In the followings we limit ourselves to quasi-one and quasi-two-dimensional systems, since for UDW

- a. the Fermi surface nesting plays an important role in the realization of the phase,
- b. we do not have yet any well established examples in three dimensional systems.

We consider the effective (low energy) Hamiltonian given by

$$H = \sum_{\mathbf{k}, \sigma} \xi(\mathbf{k}) c_{\mathbf{k}, \sigma}^+ c_{\mathbf{k}, \sigma} + \frac{1}{2} \sum_{\substack{\mathbf{k}, \mathbf{k}', \mathbf{q} \\ \sigma, \sigma'}} V(\mathbf{k}, \mathbf{k}', \mathbf{q}) c_{\mathbf{k}+\mathbf{q}, \sigma}^+ c_{\mathbf{k}, \sigma} c_{\mathbf{k}'-\mathbf{q}, \sigma'}^+ c_{\mathbf{k}', \sigma'}, \quad (1.1)$$

where $c_{\mathbf{k}, \sigma}^+$ and $c_{\mathbf{k}, \sigma}$ are the creation and annihilation operators of electrons with momentum \mathbf{k} and spin σ , $\xi(\mathbf{k})$ is the kinetic energy of electrons measured from the Fermi energy in the normal state and $V(\mathbf{k}, \mathbf{k}', \mathbf{q})$ is the interaction between two particles. In the following we represent $V(\mathbf{k}, \mathbf{k}', \mathbf{q})$ by a separable potential

$$V(\mathbf{k}, \mathbf{k}', \mathbf{q}) = \langle |f(\mathbf{k})|^2 \rangle^{-1} f(\mathbf{k}) f(\mathbf{k}') \delta(\mathbf{q} - \mathbf{Q}), \quad (1.2)$$

where \mathbf{Q} is the nesting vector. This kind of interaction is readily obtained from the extended Hubbard model with at most nearest-neighbour interaction (on

site+nearest neighbour Coulomb interaction, exchange, pair hopping and assisted hopping terms [20]). For quasi-one-dimensional systems with most conducting or chain direction parallel to the x axis, we consider $f(\mathbf{k}) = \cos(bk_y)$ or $\sin(bk_y)$. For quasi-two-dimensional systems, we consider only d-wave density waves with $d_{x^2-y^2}$ and d_{xy} symmetry. At present, these four UDW appears to exhaust all known cases. The quasi-one-dimensional description is applicable to both α -(BEDT-TTF)₂KHg(SCN)₄ and (TMTSF)₂PF₆ [52–55], while quasi-two-dimensional UDW applies to the pseudogap phase in high T_c cuprates and in CeCoIn₅ [36–39].

Within the mean-field approximation, the Hamiltonian in Eq. (1.1) is reduced to quadratic form as

$$H = \sum_{\mathbf{k},\sigma} \left(\xi(\mathbf{k}) c_{\mathbf{k},\sigma}^+ c_{\mathbf{k},\sigma} + \Delta(\mathbf{k}) c_{\mathbf{k},\sigma}^+ c_{\mathbf{k}-\mathbf{Q},\sigma} + \bar{\Delta}(\mathbf{k}) c_{\mathbf{k}-\mathbf{Q},\sigma}^+ c_{\mathbf{k},\sigma} \right) - \sum_{\mathbf{k}} \frac{|\Delta(\mathbf{k})|^2}{V \langle |f(\mathbf{k})|^2 \rangle} \quad (1.3)$$

and $\Delta(\mathbf{k})$ obeys the self-consistency equation:

$$\Delta(\mathbf{k}) = V f(\mathbf{k}) \sum_{\mathbf{k}',\sigma} f(\mathbf{k}') \langle a_{\mathbf{k}'-\mathbf{Q},\sigma}^+ a_{\mathbf{k}',\sigma} \rangle. \quad (1.4)$$

The Hamiltonian is further rewritten as

$$H = \sum_{\mathbf{k},\sigma} \Psi_{\sigma}^+(\mathbf{k}) \left[\tilde{\xi}(\mathbf{k}) \rho_3 + \eta(\mathbf{k}) + \Delta(\mathbf{k}) \rho_1 \right] \Psi_{\sigma}(\mathbf{k}) - \sum_{\mathbf{k}} \frac{|\Delta(\mathbf{k})|^2}{V \langle |f(\mathbf{k})|^2 \rangle}, \quad (1.5)$$

where $\tilde{\xi}(\mathbf{k}) = (\xi(\mathbf{k}) - \xi(\mathbf{k} - \mathbf{Q}))/2$ and $\eta(\mathbf{k}) = (\xi(\mathbf{k}) + \xi(\mathbf{k} - \mathbf{Q}))/2$. in the following we shall take the tilde off $\tilde{\xi}(\mathbf{k})$, $\eta(\mathbf{k})$ is the imperfect nesting term. From now on we limit ourselves to UCDW for simplicity. For USDW we have to involve the Pauli spin matrices as well, though the parallel treatment is possible. Also $\Psi_{\sigma}^+(\mathbf{k})$ and $\Psi_{\sigma}(\mathbf{k})$ are spinor fields conjugate to each other, $\Psi_{\sigma}^+(\mathbf{k}) = (c_{\mathbf{k},\sigma}^+, c_{\mathbf{k}-\mathbf{Q},\sigma}^+)$. Finally the single particle Green's function or the Nambu-Gor'kov Green's function is given by

$$G^{-1}(\omega, \mathbf{k}) = \omega - \xi(\mathbf{k}) \rho_3 - \eta(\mathbf{k}) - \Delta(\mathbf{k}) \rho_1. \quad (1.6)$$

Here ρ_i 's are the Pauli matrices operating on the Nambu spinor space [56]. Then the poles of $G(\omega, \mathbf{k})$ determine the quasiparticle energies as

$$\omega = \eta(\mathbf{k}) \pm \sqrt{\xi(\mathbf{k})^2 + \Delta(\mathbf{k})^2}. \quad (1.7)$$

From this, the quasiparticles density of states follows as [20, 35]

$$\frac{N(E)}{N_0} = \text{Re} \left\langle \frac{|E - \eta(\mathbf{k})|}{\sqrt{(E - \eta(\mathbf{k}))^2 - \Delta^2(\mathbf{k})}} \right\rangle, \quad (1.8)$$

where $\langle \dots \rangle$ means the average over the Fermi surface, N_0 is the quasiparticle density of states in the normal state at the Fermi energy. When $\eta(\mathbf{k}) = 0$, all

UDW so far discussed acquire the same density of states $N(E) = N_0 g(E/\Delta)$ with

$$g(x) = \begin{cases} \frac{2}{\pi} x K(x) & \text{for } x < 1 \\ \frac{2}{\pi} K(x^{-1}) & \text{for } x > 1 \end{cases}, \quad (1.9)$$

where $K(x)$ is the complete elliptic integral of the first kind [35]. For d-wave density waves, in most cases $\eta(\mathbf{k}) = \mu$, i.e. the inclusion of a finite chemical potential as imperfect nesting suffices. Then we obtain

$$N(E) = N_0 g\left(\frac{E - \mu}{\Delta}\right). \quad (1.10)$$

For nonzero chemical potential, $N(0)/N_0 = g(\mu/\Delta) \neq 0$. This is shown in Fig. 1.1. Therefore the chemical potential produces the Fermi arc or the Fermi

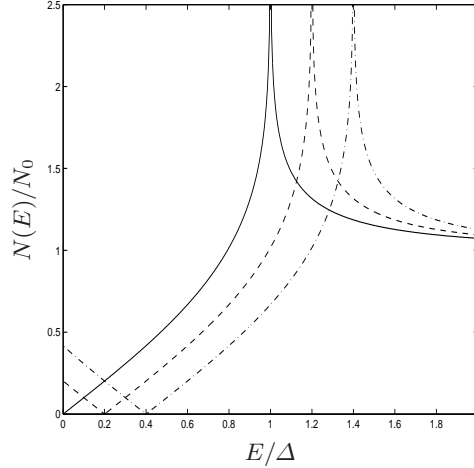


Fig. 1.1. The quasiparticle density of states of UDW is shown for $\mu/\Delta = 0$ (solid line), 0.2 (dashed line) and 0.4 (dashed-dotted line).

pockets detected in ARPES [46]. In quasi-one-dimensional systems, the difference between the different gap functions $f(\mathbf{k})$ is most readily seen in the optical conductivity $\sigma_{yy}(\omega)$ (i.e. the one with electric current parallel to the Fermi surface) [57]. The universal electric conduction (in analogy to the universal heat conduction in nodal superconductors [17]) implies that $\sigma_{yy}(0) \rightarrow 0$ as Γ and $T \rightarrow 0$ for $\Delta(\mathbf{k}) \sim \sin(bk_y)$, while $\sigma_{yy}(0) \rightarrow 2e^2 N_0 v_y^2 / \pi \Delta(0)$ for Γ and $T \rightarrow 0$ for $\Delta(\mathbf{k}) \sim \cos(bk_y)$ [58]. Here Γ is the quasiparticle scattering rate in the normal state. The Γ dependence of σ_{yy} is shown in Fig. 1.2. Note that the electric conductivity increases with quasiparticle scattering, which

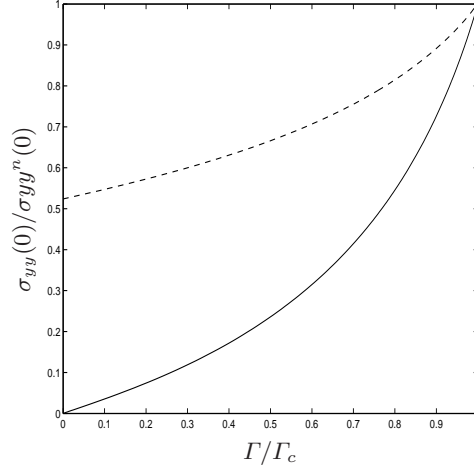


Fig. 1.2. The dc conductivity normalized by its normal state value at $T = 0$ K is plotted in the unitary scattering limit for $\Delta(\mathbf{k}) \sim \sin(bk_y)$ (solid line) and $\cos(bk_y)$ (dashed line). Γ_c is the critical scattering rate, where the DW phase vanishes. Similar curves are obtained for the Born limit.

is very counter-intuitive. But this has a similar origin to the universal heat conduction in nodal superconductors. In quasi-two-dimensional systems, the question of $d_{x^2-y^2}$ -wave density wave versus d_{xy} -wave density wave is most easily decided by the angle dependent magnetoresistance, when the magnetic field is rotated within the conducting plane as we shall discuss in Section 1.4 [37].

1.3 Landau quantization

In the presence of a magnetic field, the quasiparticle energy in UDW is quantized as first discussed by Nersisyan et al. [59,60]. The quasiparticle spectrum in the presence of magnetic field is obtained from

$$(E - \xi(\mathbf{k} + e\mathbf{A})\rho_3 - \eta(\mathbf{k} + e\mathbf{A}) - \Delta(\mathbf{k} + e\mathbf{A})\rho_1)\Psi(\mathbf{r}) = 0, \quad (1.11)$$

where we have introduced the magnetic field through the vector potential \mathbf{A} ($\mathbf{B} = \nabla \times \mathbf{A}$). It is readily recognized that Eq. (1.11) has the same mathematical structure as the Dirac equation in a magnetic field studied in 1936 [61,62]. Since the Landau quantization in quasi-one-dimensional UDW has been throughoutly discussed in Ref. [22], here we consider the Landau quantization in d-wave density waves [37]. Without loss of generality, we consider here d_{xy} -wave DW or $\Delta(\mathbf{k}) = \Delta \sin(2\phi)$. We assume that the nodal lines are perpendicular to the conducting plane and run parallel to the z axis at $(\pm k_F, 0, 0)$ and $(0, \pm k_F, 0)$. Also we take $\xi(\mathbf{k}) = v(k_{\parallel} - k_F) + \frac{v'}{c} \cos(ck_z)$,

where k_{\parallel} is the radial vector within the a-b plane, v and v' are the respective Fermi velocities in the a-b plane and parallel to the c-axis.

Let us assume that a magnetic field lies in the $z - x'$ plane and tilted by an angle Θ from the z -axis, $x' = \hat{x} \cos \phi + \hat{y} \sin(\phi)$:

$$\mathbf{B} = B(\cos(\Theta)\hat{z} + \sin(\Theta)(\cos(\phi)\hat{x} + \sin(\phi)\hat{y})). \quad (1.12)$$

We shall focus on the quasiparticle spectrum in the vicinity of Dirac cones at $(\pm k_F, 0, \pm\pi/2c)$ and $(0, \pm k_F, \pm\pi/2c)$. Then it is convenient to choose the vector potential as

$$\mathbf{A} = -B(\hat{z} \sin(\Theta) + \cos(\Theta)(\hat{x} \cos(\phi) + \hat{y} \sin(\phi)))(y \cos(\phi) - x \sin(\phi)). \quad (1.13)$$

Then in the vicinity of Dirac cones, Eq. (1.11) is recasted as

$$[E - \mu + eB(x \sin(\phi) - y \cos(\phi))(v \cos(\Theta) \cos(\phi) \pm v' \sin(\Theta))\rho_3 - v_2(-i\partial_y)\rho_1]\Psi(\mathbf{r}), \quad (1.14)$$

where $v_2 = 2\Delta/k_F$. From this, we obtain

$$(E_{1n\pm} - \mu)^2 = 2neBv_2|\cos(\phi)(v \cos(\Theta) \cos(\phi) \pm v' \sin(\Theta))| \quad (1.15)$$

$$(E_{2n\pm} - \mu)^2 = 2neBv_2|\sin(\phi)(v \cos(\Theta) \sin(\phi) \pm v' \sin(\Theta))| \quad (1.16)$$

and $n = 0, 1, 2, 3, \dots$

Except for the $n = 0$ state, which is nondegenerate, all other states are doubly degenerated. Also unlike in UDW in quasi-one dimensional systems, the Landau spectrum consists of four different branches. Furthermore, the chemical potential $\mu (\neq 0)$ is crucial in interpreting the magnetotransport of quasiparticles [37,39]. Finally, Eq. (1.15) gives the particle and hole spectrum

$$E_{1n-} = \mu \pm \sqrt{2neBv_2|\cos(\phi)(v \cos(\Theta) \cos(\phi) - v' \sin(\Theta))|}. \quad (1.17)$$

A similar formula can be worked out for Eq. (1.16). From these Landau spectra, the thermodynamics as well as the transport properties are readily obtained. In the following, we shall consider the angle dependent magnetoresistance, the non-linear Hall constant and the giant Nernst effect as the three hallmarks of UDW.

1.4 Angle dependent magnetoresistance (ADMR)

1.4.1 α -(BEDT-TTF)₂MHg(SCN)₄ salts with M=K, Rb and Tl

The nature of the low temperature phase (LTP) in the quasi-two-dimensional organic conductor α -(BEDT-TTF)₂MHg(SCN)₄ salts with M=K, Rb and Tl has not been understood until recently [63]. Although the phase transition is

clearly seen in magnetotransport measurements, neither charge nor magnetic order has been established [64, 65]. Moreover, the destruction of the LTP in an applied magnetic field suggests a kind of CDW rather than SDW. But the threshold electric field associated with the sliding motion of the density wave turns out to be very different from the one in CDW, but somewhat similar to that observed in SDW [66]: the threshold field increases smoothly with temperature and does not diverge at T_c . Indeed, the threshold electric field in the LTP of α -(BEDT-TTF)₂KHg(SCN)₄ [33, 34, 67] is well described in terms of imperfectly nested UCDW. More recently, striking angular dependent magnetoresistance has been detected in this material [68–71]. There have been many unsuccessful attempts to interpret this phenomenon in terms of the reconstruction of the Fermi surface.

Rather we find that Landau quantization of the quasiparticle spectrum in UDW as described in Sec. 1.3 plays the crucial role [52, 53]. Also the giant Nernst effect found in LTP of α -(BEDT-TTF)₂KHg(SCN)₄ can be described in terms of Landau quantized UCDW [72, 73]. Therefore these findings lead us to the conclusion that the LTP in α -(BEDT-TTF)₂MHg(SCN)₄ with M=K, Rb and Tl is UCDW.

Before proceeding, it is useful to check the Fermi surface of α -(BEDT-TTF)₂MHg(SCN)₄ salts as shown in Fig. 1.3 [4, 63].

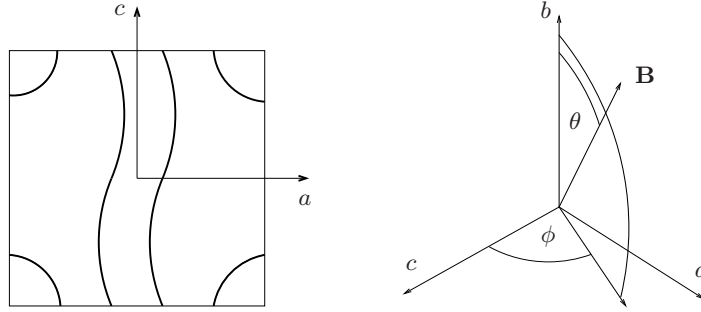


Fig. 1.3. The Fermi surface of α -(BEDT-TTF)₂KHg(SCN)₄ is shown schematically in the left panel. In the right one the geometrical configuration of the magnetic field with respect to the conducting plane is plotted.

It consists of a quasi-one-dimensional Fermi surface perpendicular to the a axis (the most conducting direction) and small two dimensional pockets (quasi-two-dimensional Fermi surface) at the corners of the Brillouin zone. Most of the de Haas van Alphen oscillations come from the quasi-two-dimensional Fermi surface, while UCDW appears on the quasi-one-dimensional one. The magnetic field configuration with respect to the conducting plane is also shown in Fig. 1.3, which was used in ADMR measurement.

The magnetoresistance is given in terms of the quasiparticle energy E_n as [22, 37]

$$R^{-1} = \sum_n \sigma_n \text{sech}^2 \left(\frac{\beta E_n}{2} \right), \quad (1.18)$$

where σ_n 's are the level conductivities weakly depending on temperature and magnetic field, $E_n = \sqrt{2nv_a \Delta e |B \cos(\theta)|}$. In the low temperature region, where $\beta E_1 \gg 1$, Eq. (1.18) is approximated as

$$\begin{aligned} R^{-1} = & 2\sigma_0 \text{sech}^2 \left(\frac{\beta E_0^1}{2} \right) + \sigma_1 \left[\text{sech}^2 \left(\frac{\beta(E_1 + E_1^{(1)})}{2} \right) + \right. \\ & + \text{sech}^2 \left(\frac{\beta(E_1 - E_1^{(1)})}{2} \right) + \text{sech}^2 \left(\frac{\beta(E_1 + E_1^{(2)})}{2} \right) + \\ & \left. + \text{sech}^2 \left(\frac{\beta(E_1 - E_1^{(2)})}{2} \right) \right] = \\ & \frac{4\sigma_0}{1 + \cosh(\zeta_0)} + 4\sigma_1 \left[\frac{1 + \cosh(x_1) \cosh(\zeta_0)}{(\cosh(x_1) + \cosh(\zeta_0))^2} + \frac{1 + \cosh(x_1) \cosh(\zeta_1)}{(\cosh(x_1) + \cosh(\zeta_1))^2} \right], \end{aligned} \quad (1.19)$$

where $\zeta_0 = \beta E_0^{(1)}$, $\zeta_1 = \beta E_1^{(2)}$ and $x_1 = \beta E_1$,

$$E_0^{(1)} = E_1^{(1)} = \sum_m \varepsilon_m \exp(-y_m), \quad (1.20)$$

$$E_1^{(2)} = \sum_m \varepsilon_m (1 - 2y_m) \exp(-y_m). \quad (1.21)$$

Here $E_0^{(1)}$ and $E_1^{(1,2)}$ are corrections to Landau level energies from imperfect nesting (i.e. the warping of the quasi-one-dimensional Fermi surface):

$$\eta(\mathbf{k}) = \sum_n \varepsilon_n \cos(2\mathbf{b}'_n \mathbf{k}). \quad (1.22)$$

From this we find $y_n = v_a b'^2 e |B \cos(\theta)| [\tan(\theta) \cos(\phi - \phi_o) - \tan(\theta_n)]^2 / \Delta c$, $\tan(\theta_n) \cos(\phi - \phi_0) = \tan(\theta_0) + n d_0$, $\tan(\theta_0) \simeq 0.5$, $d_0 \simeq 1.25$, $\phi_0 \simeq 27^\circ$ [22]. A typical fitting of ADMR of α -(BEDT-TTF)₂KHg(SCN)₄ at $T = 1.4$ K and $B = 14$ T is shown in Figs. 1.4 and 1.5.

Broad peaks centered at $\theta = 0^\circ$ comes from E_1 , while the series of dips stems from the imperfect nesting term. From these fittings, we can extract Δ (17K), $v_a \sim 6 \times 10^6$ cm/s, $b' \sim 30 \text{\AA}$, $\varepsilon_0 \sim 3$ K and σ_2/σ_1 of the order of 1/10. Finally we show in Fig. 1.6 a series of ADMR data, when the magnetic field is rotated in the plane as well. Clearly, the theory reproduces the general features of the ADMR. Of course, we notice that some interesting details are still missing from our model. Nevertheless we may conclude that UCDW describes many features of ADMR observed in α -(BEDT-TTF)₂KHg(SCN)₄ [52, 53].

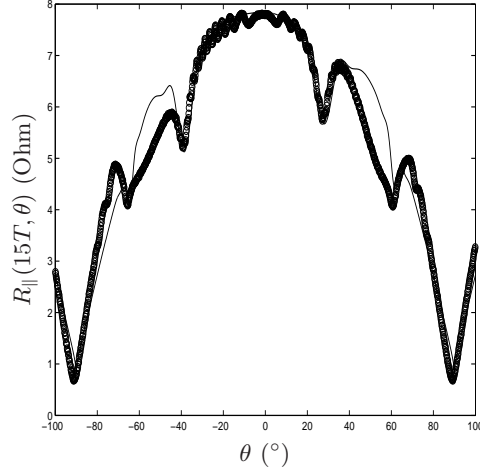


Fig. 1.4. The angular dependent magnetoresistance is shown for current parallel to the a-c plane at $T = 1.4\text{K}$, $B = 15\text{T}$. The open circles belong to the experimental data, the solid line is our fit based on Eq. (1.19).

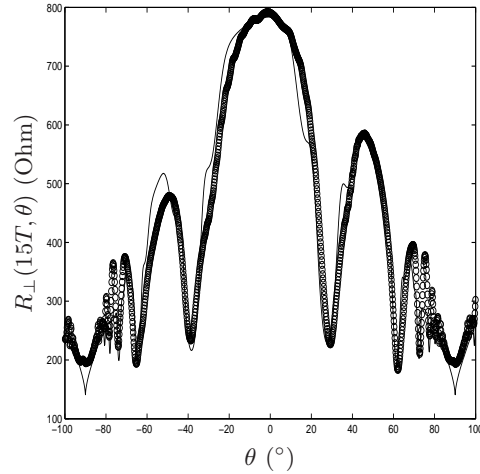


Fig. 1.5. The angular dependent magnetoresistance is shown for current perpendicular to the a-c plane at $T = 1.4\text{K}$, $B = 15\text{T}$. The open circles belong to the experimental data, the solid line is our fit from Eq. (1.19).

1.4.2 Bechgaard salts $(\text{TMTSF})_2\text{X}$

The first organic superconductor $(\text{TMTSF})_2\text{PF}_6$, discovered in 1980 [10], is, perhaps, one of the most fascinating electron system so far studied. The Bechgaard salts are well known for the variety of their ground states. They exhibit spin density wave at ambient to moderate pressure, field induced spin density wave and triplet superconductivity at high pressure ($p > 8\text{ kbar}$) [2,4,

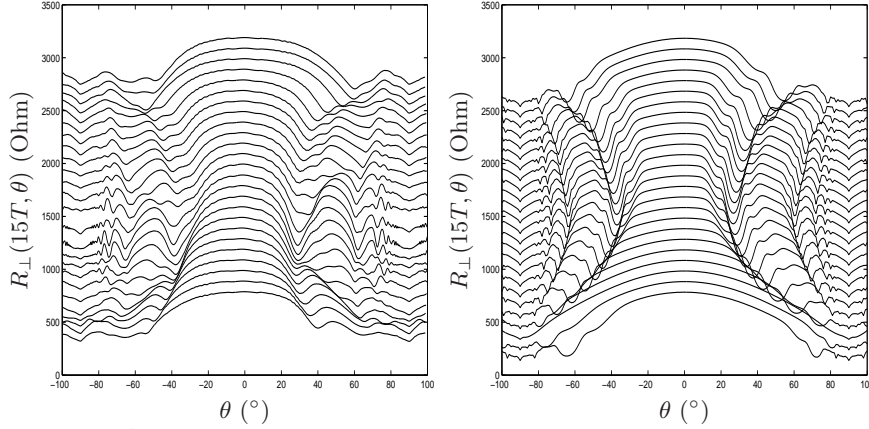


Fig. 1.6. ADMR is shown for current perpendicular to the a-c plane at $T = 1.4\text{K}$ and $B = 15\text{T}$ for $\phi = -77^\circ, -70^\circ, -62.5^\circ, -55^\circ, -47^\circ, -39^\circ, -30.5^\circ, -22^\circ, -14^\circ, -6^\circ, 2^\circ, 10^\circ, 23^\circ, 33^\circ, 41^\circ, 48.5^\circ, 56^\circ, 61^\circ, 64^\circ, 67^\circ, 73^\circ, 80^\circ, 88.5^\circ, 92^\circ$ and 96° from bottom to top. The left (right) panel shows experimental (theoretical) curves, which are shifted from their original position along the vertical axis by $n \times 100\text{Ohm}$, $n = 0$ for $\phi = -77^\circ$, $n = 1$ for $\phi = -70^\circ, \dots$

74] as shown in Fig. 1.7. We have discovered recently the coexistence of SDW

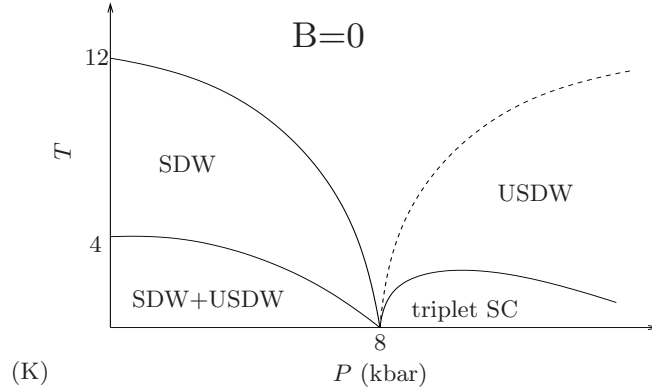


Fig. 1.7. The schematic $P - T$ phase diagram of Bechgaard salts.

and USDW at $T = T^* \sim 4\text{ K}$ in $(\text{TMTSF})_2\text{PF}_6$ during the analysis of ADMR across T^* [75]. Also in the large area of the $P - B$ phase diagram, where both SDW and superconductivity are suppressed by pressure and magnetic field (the so-called metallic state), surprising ADMR has been observed about a decade ago [76]. These are of three kind: first, when the magnetic field is rotated within the $c^* - b'$ plane perpendicular to the a axis, R_{xx} and

R_{zz} exhibits broad peaks with a number of dips [77–80]. These dips were interpreted in terms of Lebed resonance [81, 82]. More recently, very similar ADMR has been seen in the ReO_4 and ClO_4 compounds as well, though the one in the latter is more complex [54]. Second, when the magnetic field is rotated within the $a - c^*$ plane, R_{xx} show very different ADMR, sometimes called "Danner resonance" [83]. Finally the third angular dependence appears as a kink when the magnetic field is rotated within the $a - b'$ plane.

Here we shall concentrate on the case when the magnetic field is rotated within the $c^* - b'$ plane. We propose that the metallic phase should be USDW [55]. When \mathbf{B} in the $c^* - b'$ plane is tilted by ϕ from the c^* axis, the obtained energy spectrum turns out to be very similar to the one discussed in Eq. (1.17). In particular, we get $E_1 = \sqrt{2nv_a\Delta e|B\cos(\phi)|}$, $y_m = v_ad'^2e|B\cos(\phi)|(\tan(\phi) - \tan(\phi_m))^2/\Delta b$ and $\tan(\phi_m) = \frac{pb\sin(\gamma)}{qc\sin(\beta)\sin(\alpha^*)} - \cot(\alpha^*)$, where b , c , β and γ are lattice parameters in real space, α^* is a lattice parameter in reciprocal space, p and q are small integers [55]. Then the experimental data [54] of R_{xx} on $(\text{TMTSF})_2\text{PF}_6$ and on $(\text{TMTSF})_2\text{ReO}_4$ at $T = 1.55$ K are shown in Figs. 1.8 and 1.9, respectively. Except for the fact that the theoretical curves exhibit more structures than the data, we think the fitting is excellent. From these, we extract $\Delta = 20$ K and 45 K and $v_a = 10^7$ cm/s and 3×10^7 cm/s for the PF_6 and ReO_4 compound, respectively. The Fermi velocities deduced here are also very consistent with known values. Also from the above Δ 's, we expect that USDW persists to $T = 9$ K and 20 K for the respective compounds. Also the giant Nernst effect in these systems can be crucial to strengthen the case of USDW in Bechgaard salts [84].

1.4.3 κ -(ET) $_2$ salts, CeCoIn_5 and YPrCO

There are three κ -(ET) $_2$ salts with very similar properties: κ -(ET) $_2\text{Cu}(\text{NCS})_2$, κ -(ET) $_2\text{Cu}[\text{NCN}]_2\text{Br}$ and κ -(ET) $_2\text{Cu}[\text{NCN}]_2\text{Cl}$ with relatively high superconducting transition temperature $T_c \gtrsim 10$ K. So far no evidence for unconventional density wave in these systems has been reported, although we have many reasons to believe that the pseudogap phase in high quality crystals should be d-wave density wave. First of all, There are many parallels between these organic superconductors and high T_c cuprate superconductors and the heavy fermion compound CeCoIn_5 :

- quasi-two-dimensionality (or layered structure),
- the proximity of antiferromagnetic order,
- d-wave superconductivity.

Of course d-wave superconductivity is established only for κ -(ET) $_2\text{Cu}(\text{NCS})_2$ [85], but this suggests strongly that the other two κ -(ET) $_2$ salt superconductors should be d-wave as well. Second, the giant Nernst signal and ADMR observed in the pseudogap phase of high T_c cuprates [40–43] and in CeCoIn_5 [39, 44] are successfully interpreted in terms of d-wave density wave [37]. These suggest strongly the presence of dDW in κ -(ET) $_2$ salts.

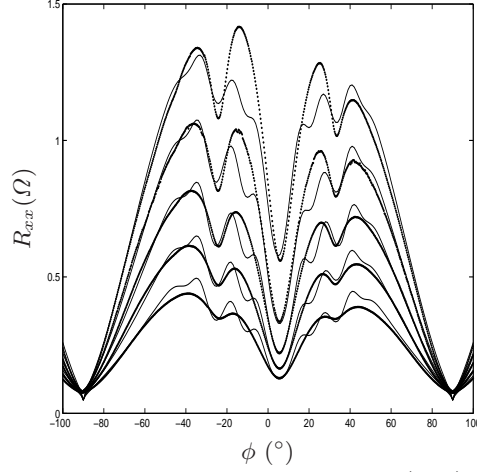


Fig. 1.8. The angular dependent magnetoresistance (R_{xx}) of $(\text{TMTSF})_2\text{PF}_6$ at $T = 1.55$ K is shown for magnetic fields from 8-4 T from top to bottom. The dots denote the experimental data from [54], the solid line is our fit based on equation (1.19).

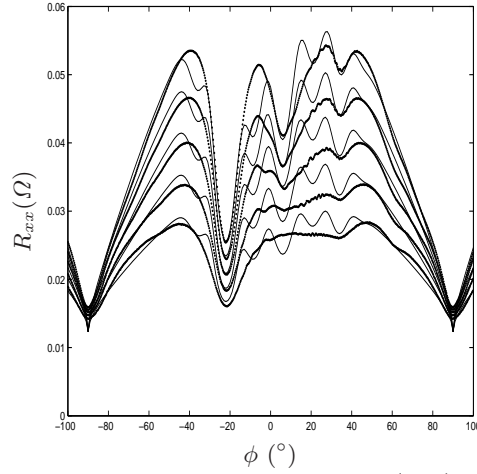


Fig. 1.9. The angular dependent magnetoresistance (R_{xx}) of $(\text{TMTSF})_2\text{ReO}_4$ at $T = 1.55$ K is shown for magnetic fields from 8-4 T from top to bottom. The dots denote the experimental data from [54], the solid line is our fit based on equation (1.19).

Here we are going to show the fitting of ADMR in the pseudogap phase of CeCoIn_5 [39] and in high T_c cuprate $\text{Y}_{0.68}\text{Pr}_{0.32}\text{Ba}_2\text{Cu}_3\text{O}_7$ [37, 43] in Figs. 1.10 and 1.11, respectively.

From these fittings, we can extract $\Delta = 45$ K and 360 K, μ (the chemical potential)=8.4 K, 40-60 K, v (the planar Fermi velocity)= 3.3×10^6 cm/s and

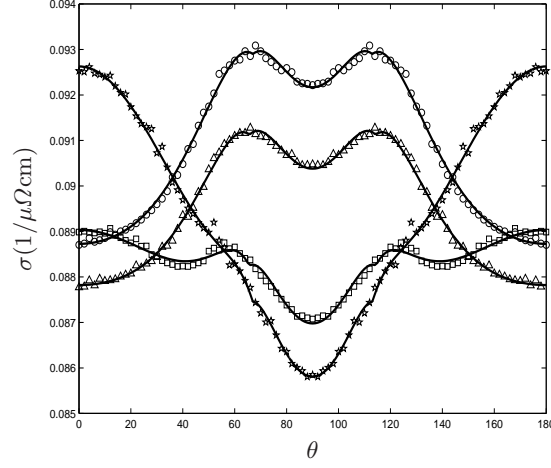


Fig. 1.10. The angle dependent conductivity of CeCoIn₅ is shown for $T = 6$ K and for $B = 4$ T (circle), 5 T (triangle), 8 T (square), 10 T (star).

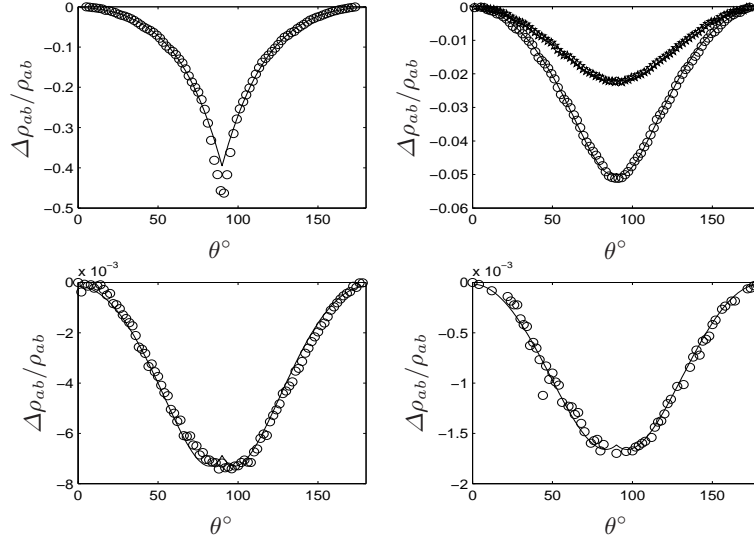


Fig. 1.11. The relative change of the in-plane magnetoresistance of Y_{0.68}Pr_{0.32}Ba₂Cu₃O₇ [43] is plotted as a function of angle θ at $B=14$ T for $T=52$ K (top left), 60 K and 65 K (top right), 75 K (bottom left) and 105 K (bottom right). The solid line is fit based on Eq. 1.19.

2.3×10^7 cm/s for CeCoIn₅ and YPrCO, respectively. These values are very consistent with known parameters of these systems.

1.5 Giant Nernst effect

Since 2001 the giant Nernst effect has been established as the hallmark of the pseudogap phase in high T_c cuprates [40–42]. As shown in Ref. [22, 36], the giant negative Nernst effect follows directly from the Landau quantization of the quasiparticle energy spectrum in UDW. When an electric field is applied in the conducting plane in addition to the perpendicular magnetic field, all quasiparticles drift with a drift velocity $\mathbf{v}_D = \mathbf{E} \times \mathbf{B}/B^2$. This gives rise to a heat current $\mathbf{J}_h = TS\mathbf{v}_D$, where S is the quasiparticle entropy given by

$$S = eB \sum_n \left\{ \ln(1 + e^{-\beta E_n}) + \beta E_n (1 + e^{\beta E_n})^{-1} \right\}, \quad (1.23)$$

where the sum has to be carried out over all the Landau levels. For d-wave density wave as in high T_c cuprates and for $\mathbf{B} \parallel \mathbf{c}$ and $\beta E_1 \gg 1$, the entropy is well approximated as

$$S = 8eB \left[\ln(1 + e^{-\zeta_0}) + \zeta_0 (1 + e^{\zeta_0})^{-1} + \ln(2(\cosh(x_1) + \cosh(\zeta_0))) - \frac{\zeta_0 \sinh(\zeta_0) + x_1 \sinh(x_1)}{\cosh(x_1) + \cosh(\zeta_0)} \right], \quad (1.24)$$

where $\zeta_0 = \beta\mu$, $x_n = \beta\sqrt{2neBv_2v}$ with $v_2/v = \Delta/E_F$. Then the Nernst coefficient is given by

$$S_{xy} = -\frac{S}{B\sigma} = \frac{1}{\sigma} \left\{ \frac{L_{2D}}{1 + (B/B_0)^2} - 8e \left(\frac{1}{2} \left[\ln(2(1 + \cosh(\zeta_0))) - \frac{\zeta_0 \sinh(\zeta_0)}{1 + \cosh(\zeta_0)} \right] + \ln(2(\cosh(x_1) + \cosh(\zeta_0))) - \frac{\zeta_0 \sinh(\zeta_0) + x_1 \sinh(x_1)}{\cosh(x_1) + \cosh(\zeta_0)} \right) \right\}. \quad (1.25)$$

Here σ has been already defined in Eq. (1.18), and L_{2D} and B_0 comes from quasiparticles not in d-DW state. Here we present our fitting to the data taken on α -(BEDT-TTF)₂KHg(SCN)₄ [72] in Fig. 1.12.

We have also similar fittings for high T_c cuprates [36] and CeCoIn₅ [38]. The third hallmark of UDW is the nonlinear Hall coefficient. In UDW in an applied magnetic field, the number of quasiparticles decreases exponentially with decreasing temperature. Therefore the Hall conductivity is given by [47]

$$\sigma_{xy} = -\frac{2e^2 \cos^2(\Theta)}{\pi} n(B, T) \quad (1.26)$$

with

$$n(B, T) = \tanh(\zeta_0/2) + \sinh(\zeta_0) \left[\frac{1}{\cosh(x_1) + \cosh(\zeta_0)} + \frac{1}{\cosh(x_2) + \cosh(\zeta_0)} \dots \right], \quad (1.27)$$

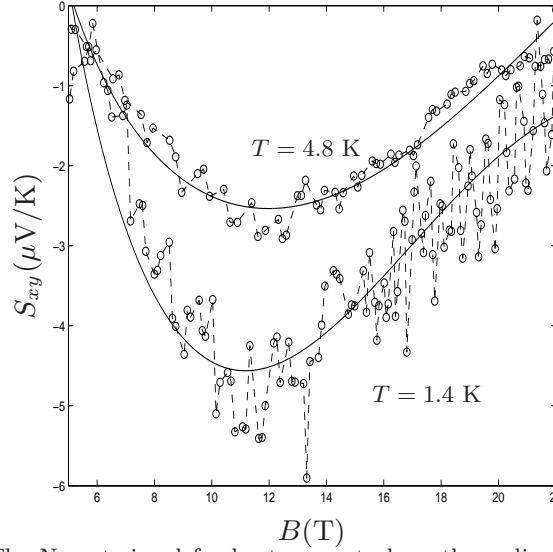


Fig. 1.12. The Nernst signal for heat current along the a direction is shown for $T = 1.4$ K and $T = 4.8$ K (from bottom to top), the dashed lines with circles denote the experimental data from Ref. [72], the solid line is our fit based on Eq. (1.25).

where Θ is the angle the magnetic field makes from the c axis. We have no possibility to compare the theoretical expression (Eq. (1.26)) to available experimental data. But clearly the Hall coefficient will indicate the reduction of the number of quasiparticles due to the opening of the energy gap.

1.6 Concluding remarks

In the last few years we have experienced a major paradigm shift from conventional condensates to unconventional condensates. These are still the mean-field ground states and described in terms of the generalized BCS theory. For superconductivity, at least d-wave superconductivity in κ -(ET)₂ salts and triplet superconductivity in (TMTSF)₂PF₆ (most likely chiral f-wave) has been established [17,86]. As to density waves, more and more previously unidentified condensates appear to belong to UDW. But this game has just started. Also there appears to be still unexplored areas where two of the mean-field order parameters can coexist. Of course, the classic example is NbSe₃ where the CDW's: CDW₁ and CDW₂ coexists. A more exotic case will be in Bechgaard salts (TMTSF)₂PF₆ where SDW and USDW appears to coexist below $T^* \sim T_c/3$ [75]. However, Gossamer superconductivity appears to be more widespread, where a nodal superconductor coexists with a nodal density wave [47,87]. Actually this is our interpretation of the concept of Gossamer superconductivity introduced by Bob Laughlin. As discussed in

Ref. [47], the case of Gossamer superconductivity in high T_c cuprate superconductors, CeCoIn_5 and very likely $\kappa\text{-(ET)}_2$ salts is clear [88].

In other words all the electronic ground states in crystalline solids belong to one of three possibilities:

- a. unconventional superconductivity
- b. unconventional density wave and
- c. coexistence of two of them.

These ground states have been expected from the renormalization group analysis of 2D and 3D fermion systems. In these circumstances, the identification of the character and the symmetry of the order parameter is the first step. Now with ADMR, the giant Nernst effect and the nonlinear Hall coefficient, the exploration becomes much easier.

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